PATENT SPECIFICATION

NO DRAWINGS

Inventors: HIROSUKE YUMOTO, KAZUYA HARADA and MASAAKI ITOGA

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COMPLETE SPECIFICATION

Improvements in or relating to Polyethylene Terepthalate Films

We, TOYO RAYON KABUSHIKI KAISHA, a corporation organized under the laws of Japan, of No. 2, 2-chome, Nihonbashi-Muromachi, Chuo-ku, Tokyo, Japan, do hereby declare the invention, for which we we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to polyethylene terephthalate film and is concerned, more particularly, with a method of improving the dimensional stability of polyethylene terephthalate films.

It is known that a thermoplastic film which has been stretched longitudinally and transversely can be subjected to a heat treatment with a view to improving its dimensional stability. However, it is difficult to obtain a polyethylene terephthalate film having improved dimensional stability by the known methods, and a film prepared by such methods generally possesses a thermal shrinkage of more than 2 or 3% and, therefore, cannot be used in applications which require film having a highly stabilized shape, for example insulators for electrical equipment, and recording tape.

It is an object of the present invention to provide a method whereby the disadvantage mentioned above can be substantially overcome and polyethylene terephthalate film of improved dimensional stability may be obtained.

Accordingly, the invention provides a method of improving the dimensional stability of a polyethylene terephthalate film, which comprises subjecting a polyethylene terephthalate film, which has been stretched longitudinally and transversely, to a first heat treatment at a temperature in the range of from 150 to 250°C. whilst maintaining the film under tension and its longitudinal and transverse dimensions substantially constant, and thereafter subjecting the heat-treated polyethylene terephthalate film to a second heat treatment at a temperature in the range of from 150 to 250°C. whilst allowing the film to be relaxed along its longitudinal and/or transverse dimensions by from 2 to 30%.

The method of the present invention enables polyethylene terephthalate film having a thermal shrinkage of not more than 1% to be prepared. The thermal shrinkage of a polyethylene terephthalate film can be determined as follows: a piece of film having a length of 15 cm. and a width of 1 cm. is taken and two lines 10 cm. distant from each other marked on it. From the lower end of the film a weight of 1 g. is suspended when a film of 12 μ thickness is employed. When a film having a thickness other than 12 μ is employed, the weight employ is such that the force per unit cross-sectional area of the film is the same as when a film having the cross-section of 1 cm. $\times$ 12 μ is employed. The specimen is kept at 150°C. in air and the decrease in distance between the two marked lines is measured after one minute by means of a cathetometer. The percentage decrease in length over the original length is calculated and represents the thermal shrinkage.

Through many experiments, it has now been found that although internal stresses remain in a polyethylene terephthalate film which has been stretched and oriented in two directions and subjected to a single heat treatment whilst under tension or whilst relaxed, such stresses can be removed and the dimensional stability at high temperature can be improved when the film is subjected, in accordance with the invention, to

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at least one further heat treatment. The first heat treatment serves to fix the orientation caused by the stretching. If the first heat treatment is not made, the orientation caused by the stretching is decreased by the relaxation and an improvement in the dimensional stability of the film becomes difficult to obtain. However, when a polyethylene terephthalate film which has been subjected to a first heat treatment, in which the molecular orientation caused by stretching is fixed, is subjected to a second heat treatment in which the film is allowed to be relaxed in one direction and/or in the direction perpendicular thereto. the internal stresses remaining after the first heat treatment can be effectively removed and the thermal shrinkage decreased considerably. Advantageously, the second heat treatment is carried out at a temperature which is equal to or higher than the temperature at which the first heat treatment is carried out.

In some cases, when a lowering or variation of temperature occurs in the final period of the second heat treatment, the desired improvement is not fully achieved and, in such cases, it is desirable to carry out a third heat treatment in order to secure the required improvement. When a polyethylene terephthalate film is subjected to such a third heat treatment the temperature employed is again in the range of from 150°C. to 250°C. The third heat treatment is carried out, as a rule, under tension. However, when a large relaxation occurs in the second heat treatment, the third heat treatment may be carried out in the condition that almost no tension exists in the transverse direction, causing occasionally the film to be slackened as described later.

When the heat treatments are carried out below 150°C. extremely long periods are required. This is not efficient. Further, when the heat treatments are performed at a considerably lower temperature than 150°C., the desired effect is not obtained. When the heat treatments are performed at a temperature above 250°C. a softening of the polyethylene terephthalate film and a lowering of the mechanical strength thereof occur.

The times necessary for the first, second and, when employed, third heat treatments are determined by the thickness of the thermoplastic film, the stretching ratio and the temperatures of the heat treatment. In most cases, the time required for the first and second heat treatments is from 3 to 30 seconds. The relaxation which occurs in the second heat treatment may be accomplished in a moment but the time necessary for the accomplishment depends on the size of apparatus. A time of more than 30 seconds makes it difficult to make the process continuous.

In the second heat treatment, there is employed a relaxation of from 2 to 30%. When a relaxation higher than 30% is employed, a slackening is formed as the film passes through the outlet of the stretching machine and in such cases the film becomes wrinkled when the treated film is wound up. The occurrence of slackening depends on the temperature of heat treatment. When a high temperature is employed, the film is not wrinkled even if a relaxation of about 30% is employed.

It can be noted, however, that even if slackening does occur and the film is wrinkled, the winding-up may still be performed by expanding the width to remove the slackening or by widening the film in the transverse direction by means of an expander-roll or the like.

In the thermal relaxation treatment of this invention, an error in fixing the dimension cannot be avoided entirely. Accordingly, the variation in the thermal shrinkage often becomes quite large. Therefore, it is preferable to employ a relaxation higher than 2%.

In practising this invention, it is preferable to employ a stretching machine of improved tenter type in which the thermoplastic film, both edges of which are gripped, is introduced into a heating equipment where the film is heated by hot air or by an infrared heater. The relaxation treatment is performed bv allowing the width of the film to narrow.

Thermoplastic film in the form of a tube can be treated in the following way. Two positions along the longitudinal direction are pinched by rollers and air is injected into it to expand the tube. Then, the expanded tube is heated by an infra-red heater or by hot air whilst the dimensions of the tube are maintained substantially constant. The film is then cooled. In the second heat treatment the film is again held by nip rollers, expanded by the injection of air and heat-treated, the film being allowed to relax in the longitudinal direction by employing different peripheral speeds for the nip rollers and/or in the transverse direction by adjusting the internal air pressure. The cooling step between the first and second heat treatment has no deleterious effect on the results obtained by application of the method.

The invention will now be illustrated by reference to the following Examples.

Example 1

Four polyethylene terephthalate films of 50 μ thickness (Samples 1, 2, 3 and 4) which had been prepared by stretching until the stretching ratios in the longitudinal and transverse directions were 3.4 and 3.6 respectively, were subjected to a heat treatment. The films under tension were placed in a first chamber at 220°C. for 8 seconds, and then subjected to a relaxation treatment in the transverse direction in which the films

were treated for 4 seconds at the same temperature until a thermal relaxation of 1% (Sample 1), 7% (Sample 2), 10% (Sample 3) and 15% (Sample 4) was obtained. The resulting films were then transferred to a second chamber at a temperature of 220°C. for a third heat treatment for 11 seconds without dimensional charge.

The thermal shrinkage of these four samples was found to be as shown in the following table.

TABLE 1

	Sample 1	Sample 2	Sample 3	Sample 4
Transverse direction	1.8—2.8%	0.9—1.1%	0.3—0.5%	0
Longitudinal direction	1.7—2.0%	1.2—1.3%	1.1—1.2%	1.0—1.1%

Comparative Example

Two polyethylene terephthalate films of 50 μ thickness (Samples 5 and 6) which had been prepared by stretching until the stretching ratios in the longitudinal and transverse directions were 3.4 and 3.6 respectively, were subjected to a heat treatment with relaxation in which the films at a temperature of 180° C. were allowed to be relaxed for 4 seconds until a relaxation of 7% (Sample 5) and 10% (Sample 6) was obtained. The resulting films were then transferred to a second chamber having a temperature of 200°C. for heat treatment and kept there for 11 seconds. The thermal shrinkage of these two samples was found to be as shown in the following table.

TABLE 2

	Sample 5	Sample 6
Londgitudinal direction	2.1%	2.0%
Transverse direction	3.0%	2.8%

WHAT WE CLAIM IS:—

1. A method of improving the dimensional stability of a polyethylene terephthalate film, which comprises subjecting a polyethylene terephthalate film, which has been stretched longitudinally and transversely, to a first heat treatment at a temperature in the range of from 150 to 250°C. whilst maintaining the film under tension and its longitudinal and transverse dimensions substantially constant, and thereafter subjecting the heat-treated polyethylene terephthalate film to a second heat treatment at a temperature in the range of from 150 to 250°C. whilst allowing the film to be relaxed along its longitudinal and/ or transverse dimensions by from 2 to 30%.

2. A method according to Claim 1, wherein the second heat treatment is carried out at a temperature which is equal to or higher than the temperature at which the first heat treatment is carried out.

3. A method according to Claim 1 or 2, wherein the polyethylene terephthalate film is subjected to a third heat treatment at a temperature in the range of from 150 to 250°C.

4. A method according to Claim 1, 2 or 3, wherein in said second heat treatment the film is relaxed by an amount of from 2% to 10%.

5. A method according to Claim 1, 2, 3 or 4, wherein the film is subjected to each heat treatment for from 2 to 30 seconds.

6. A method according to Claim 1, substantially as described in the foregoing Example 1.

7. Polyethylene terephthalate film, the dimensional stability of which has been improved by the method claimed in any one of the preceding claims.

HASELTINE, LAKE & CO.,
Chartered Patent Agents,
28 Southampton Buildings,
Chancery Lane,
London, W.C.2.
Agents for the Applicants.

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